Final Report

COOPERATING TO IMPLEMENT THE GREAT LAKES WATER QUALITY AGREEMENT
MISE EN OEUVRE DE L'ACCORD SUR LA QUALITÉ DE L'EAU DES GRANDS LACS

Technical Review of the

Integrated Atmospheric Deposition Network (IADN)

- Technical review of monitoring and research activities of the IADN program (1990-2002)
- Recommendations for the future operation and direction of the network.

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Preamble

Annex 15 of the Great Lakes Water Quality Agreement sets out actions to be taken by the Parties with the general intent of reducing atmospheric deposition of toxic substances in the Great Lakes Basin. *IADN satisfies some, but not all, of the requirements*. It does not, for example, address effects or pollution control measures. The IADN related focus is on loadings and temporal and spatial trends in concentrations.

It was, and is, recognized that the program should evolve to take advantage of new developments and especially improvements in methods of chemical analyses, methods of estimating loadings, and methods to determine source-receptor relationships. Some particular research activities were suggested in the Annex with a view to effecting these improvements, but as knowledge is gained, as concentrations change, as new chemicals of concern are identified and the significance of atmospheric and other pathways becomes clearer, these research activities and areas for improved practices must change; otherwise the programs will stagnate and become less relevant to the needs of the residents of the Great Lakes Basin. Indeed the Review Panel questions if "research" is the best descriptor of a set of activities which can broadly be described as simply "doing things better" and ensuring that the program operates at the current "state of the science".

Much of the effort has been devoted to improving the accuracy of chemical analyses of chemicals in the atmosphere and in precipitation. It is essential that such efforts continue with the objective of ensuring that IADN remains an internationally recognized monitoring program with a more than 10-year continuous record of data. It must be appreciated, however, that efforts must continue to improve methods of calculating loadings from concentrations and as the Annex states determining "the significance of atmospheric loadings to the Great Lakes System relative to other pathways" and "the source of such (toxic) substances…." The latter efforts are an integral part of the IADN program, and have received inadequate attention to date.

Accordingly, in this report the Panel comments on the program of chemical analyses, on the tasks of establishing temporal and spatial trends in concentrations, and it makes recommendations for improvements, including the addition of new substances. It also comments on tasks of determining loadings and their uncertainties on a whole lake basis, on a Basin-wide basis and taking into account local urban influences, as well as contributing to establishing a clearer "big picture" of contaminant sources and pathways to and within the Basin. Specific suggestions are made to address perceived weaknesses in the calculation of loadings and to better identifying the key sources of uncertainty.

Key Recommendations in IP3:

Are the future recommendations in the technical summary appropriate given our current understanding of atmospheric deposition of toxic chemicals to the Great Lakes? Are there still important questions going unanswered that could be addressed by IADN?

1. **Scientific Advisory Panel**: Recognizing the need to remain current and connected to international organizations, IADN should consider establishing an external scientific advisory group and/or steering committee of broad scientific expertise to advise IADN and Parties on a *regular* (perhaps annual) basis.

2. **Mass Balancing**: The Parties should support an effort to re-visit the big-picture view of mass balancing of chemicals in the Great Lakes in order to re-focus IADN and the community to the role of atmospheric loading to the Great Lakes (e.g., Strachan and Eisenreich Report, Niagara on the Lake Report). This effort would also focus the overall challenge of assessing key data gaps with regard to the question of relative loadings.

3. **Urban Strategy**:
    a. IADN and the Parties should assess how urban emissions affect lake-wide loading estimates in a scientifically credible manner.
    b. This question should be viewed in the larger context of the overall problem of determining source-receptor relationships for each pollutant of concern for each lake. It is crucial to know the relative importance of different source areas and source types. The urban strategy should include IADN sampling and other urban monitoring efforts (PAHs, Hg, PM2.5, etc) and strategic research on *measurement* and *modeling* in urban airsheds and their immediate vicinity. The use of the urban IADN sites as a platform will facilitate gradient studies and innovative tools such as passive sampling
    c. IADN should evaluate the utility of urban sampling efforts to contribute to improved loading estimates. The current urban monitoring schemes may not be particularly useful in generating more accurate loading estimates, due to large and unknown spatial gradients.
    d. Estimation of loading due to emissions in urban areas will most likely require a combined approach involving both modeling and monitoring. The needs of the overall modeling-assisted loading estimation process should be considered in the design and implementation of any urban monitoring program.
    e. Since monitoring budgets are limited and the utility of urban measurements to assist in loading estimates is uncertain, consider the use of a mobile platform that could be utilized periodically in a number of different urban areas, serving primarily as a screening tool. Such a mobile platform might also be used to help characterize spatial and temporal variations in pollutant concentrations, and provide evaluation or calibration data for models. IADN should promote the use of passive samplers in urban areas, at IADN sampling

sites and at other localities with a view to increasing spatial coverage with minimal additional cost .

4. **Improving loading estimates to Great Lakes**:
    a. IADN should consider only downward loading estimates in total loading lake estimates: wet deposition, dry particle deposition, and gross gas absorption. This provides the clearest picture of the magnitude and importance of air-water exchange. This also avoids uncertainties and ealys associated with obtaining water concentrations. This is not to say that volatilization (or water to air exchange) is not important; the panel simply recognizes that absorption is deposition, and volatilization is a loss term.
    b. IADN scientists should review up to date scientific and process information on dry particle deposition, wet deposition and gross gas deposition prior to making loading estimates, and implement improved estimate methodologies after review by an expert panel in order that current science is fully exploited.
    c. IADN should improve its simplifying assumptions in loading calculations– less aggregation of meteorological data and water temperatures; less aggregation of concentration data. Seasonally averaged water and air temperatures and wind speeds are insufficient. Integration of meteorological data in RDMQ should be combined with concentration data and RDMQ should be used to calculate loadings estimates directly.
    d. IADN should recognize that improved loading estimates requires modeling expertise given spatial and temporal variations, and the fact that the monitoring is necessarily very limited, it will not generally be possible to create accurate loading estimates from monitoring alone.
    e. IADN should establish a strategy to assess the importance of atmospheric deposition to the Great Lakes Basin, assessing the loading of connecting channels to the down gradient lakes that result from atmospheric deposition to the up-gradient lake. Additionally, consideration should be given to developing methods for assessing atmospheric depsotion to the terrestrial watersheds of the Great Lakes, and that fraction of which that reaches the Lakes.
    f. IADN should assess the adequacy of the intermittent sampling strategy now being used for ambient air concentrations, evaluating the significance of temporal variations in generating uncertainties in loading estimates.

5. **Chemicals: Nominations, Listing, De-listing, Emerging Chemicals**

As noted on p. 15 of the IADN Technical Summary, the nomination and consultation process has not been successful. The availability of the nomination process to the scientific community, the policy makers and the public is not widely appreciated, and there may be skepticism that nominations will be considered seriously. Preparing a full chemical dossier is a demanding task.

The Review Panel recommends that the nomination process be re-visited to facilitate nominations. The availability of the nomination process should continue to be widely publicized. We suggest the establishment of a two tier system with a first tier as an open

"clearing house" containing a large number of suggestions from a variety of sources, and a second more selective list of substances which are being assessed in more detail for analytical feasibility and human and ecosystem effects.

A biannual report on the nominations, assessments and listing/de-listing of chemicals should be prepared. The following substances should be considered for nominations: Hg, PBDEs, chlorinated alkanes, chlorinated naphthalenes, polyfluorinated substances, PCDDs/Fs, and current use pesticides and herbicides. Regarding the latter, a useful initial step would be to survey the current pesticide application practices and rates in the Great Lakes Basin, thus providing a perspective on candidate substances. The list of target chemicals should be linked to the priority chemical lists active in Europe and Japan.

Additional comments on chemical listing and delisting:

a. Evaluate how best to introduce Hg deposition to IADN: Hg deposition network, new real-time methodologies for reactive, gaseous, and particulate Hg; perhaps focus on Hg for some years.
b. Consider adding metals to all master sites with Canada performing analyses.
c. Consider doing organic chemical analyses for specific analytes at a single laboratory for all sites (e.g., PBDEs at IU; atrazine at MSC; PCDDs/Fs at IU, current use herbicides at MSC). This should result in improved precision.
d. Continue pro-active analysis of potential emerging chemicals.
e. Consider de-listing or substantially reducing the effort for "old" organochlorine pesticides on the list of target analytes.
f. Listing or Delisting should depend on ecological and/or human health significance, not only on analytical capability.

6. **International Linkages**: Link IADN to other national and international monitoring and research networks in North America and Europe (especially EMEP). The problem of atmospheric transport and deposition of POPs and other contaminants is not unique to the Great Lakes region of North America. Linkages to key international efforts in this area will raise the profile of IADN, improve the exchange of valued information, and lead to important considerations of cross-boundary transport and deposition.

7. **Laboratory Analyses**: It is essential that the IADN Management Team implement immediate improvements in the quality and timeliness of analytical measurements and reporting. The Panel recommends that serious consideration be given to focusing some analyses in Canadian laboratories and others in the Indiana University or EPA contract laboratories. Consider dropping PCB analyses in precipitation at the Canadian sites OR having all PCB analyses performed at the Indiana University laboratory.

International Perceptions of IADN and its Achievements

IADN is one of the few integrated, long-term networks for monitoring of persistent trace substances in the world. It generally yields data of high quality and it is an example of a successful scientific partnership between two countries - Canada and the US.

IADN was amongst the first air toxics monitoring networks to be established globally; it is yielding invaluable long-term trend data for a variety of priority pollutants. There is an impressive continuity to the trend record, at a range of locations. These datasets are not only important nationally, but they are also important to the wider international community through UNEP and other organizations.

The focus of the data reaching the international community is POPs in air. Key publications highlight spatial and temporal trends, and improve our understanding of the sources and environmental processes that control ambient levels and ecosystem exposures to a range of substances. There is a strong emphasis on the analysis of data from the US sites in the peer-reviewed literature, most frequently because of the later release of data on the Canadian side and less motivation to publish the results of the network in the scientific literature. As a result, the perception is growing that there is a lack of commitment in Canada to data analysis, interpretation and publication, especially in the last few years. A more vigorous Canadian scientific effort is essential.

The monitoring and surveillance programs provide a unique perspective on the complexities of the processes affecting the Great Lakes ecosystems. IADN's primary goal - to quantitatively assess the role of atmospheric deposition to the Great Lakes basin - is well designed to inform management and decision-making for those key water bodies. The new IADN management team has made important strides during IP2 to improve the accessibility and "profile" of data to the public and the wider scientific community. It is noted that, unlike monitoring programs such as the Mercury Deposition Network, detailed monitoring data are not yet available on the IADN Website. Interested parties must submit a request for the data. While accessibility to the data has improved, there is still room for progress, e.g., the provision of key datasets directly through the Internet. The improved data management and QA/QC procedures are very beneficial and will serve as excellent examples of how other monitoring programs should be structured.

Compared to other international networks on atmospheric deposition such as AMAP, EMAP, OSPAR (North Atlantic) and HELCOM (Baltic Sea), IADN is exceptional in the very high focus given to the monitoring of POPs. EMEP is the largest network in Europe, focusing traditionally on transboundary sulfur and nitrogen. POPs and trace metals were not included in EMEPs monitoring programme before 1999. EMEP has, however, collected data on POPs and trace metals among the participants (mostly AMAP, OSPAR and HELCOM data) since 1995. EMEP has provided almost equal focus to trace metals and POPs, and on measurements and modelling, respectively. EMEP has sampling/analysis guidelines and QA programs for POPs and trace metals. OSPAR and HELCOM have some, whereas EMAP does not have harmonized guidelines/ QA protocols.

Some identified problems and suggestions for improvement

While IADN has demonstrated considerable success in accomplishing its mission, that mission has narrowed remarkably from the initial vision for IADN covered by the drafters of Annex 15. IADN was formulated as a "research, surveillance and monitoring network" and it has become increasingly only a monitoring"" network. The interest in staying relevant and current will be a problem unless the IADN program can step back and understand what it needs to do to better to address Annex 15 goals. The measurement of air and precipitation concentrations are, by and large, a successful effort under the program; however, the improvement on estimations of overall loadings to the basin, the determination of the relative importance of different loading pathways, and the determination of the relative importance of different sources within any loading pathway have not progressed significantly. The "loading paradigm" needs to be re-examined, probably through a modern version of the workshop leading to the Strachan and Eisenreich report) from which a new loading paradigm may evolve.

Problems the Panel has identified are:

1. **Lack of improvement in the "integration" component of the Network**. . There is a tension between laboratories and agencies that does not serve to improve the overall product. The QA program has identified a number of problems (e.g., 2:1 or 3:1 differences in PCB measurements in air; 10:1 differences in PCB measurements in precipitation). These need to be resolved and, if one sampling methodology/laboratory combination cannot adequately make reliable measurements, the management team should change the sampling methodology and/or laboratory (whichever is needed) to one with a proven record.

2. **Improvement in the precision of the estimation of loadings is unlikely given the current loading paradigm**. IADN should focus on the parts of the loading equation that it can measure (wet and dry deposition and gas absorption). Improvement in these three terms will be an important contribution to IP3. Areas of improvement include: estimates of the overall mass transfer coefficient(s), K_{oL}, should be matched in space and time to the air measurements; better estimation of lake surface temperature (NOAA data can be obtained on a weekly basis…this should be matched to sampling days and lake temperatures from the vicinity of the measurement sites). Are there terms in the loading paradigm that are insignificant for some compounds? If so, streamline the process of calculating loadings. Loadings should be estimated immediately from RDMQ with some, a priori, inputs. These should made available at the same time the concentration data are available. It is not necessary to wait for a loadings workshop to present this "first look" loadings estimates to the public. The loadings workshop can be a review and quality control on the routine computation of the loadings from RDMQ.

3. **Turn around time of the data, while considerably improved from IP1, is still inadequate**. The laboratory mass levels of chemicals in the atmospheric compartments should be available to RDMQ by July-August for the preceding year. These amounts should be considered "interim" and allow the Data Officer to begin quality control of the data. Revisions by the laboratory analysts will arise naturally from this process. Final quality controlled datasets of concentration and interim loadings should be available for the Loadings Workgroup not later than 12 months after last sample collection.

4. **IADN has dropped certain classes of chemicals from its Tier lists, not by scientific review but by funding issues**. Trace metals (e.g., Pb, As, Se, Cd) should be reinstated in the IADN list of compounds at all Master Sites.

5. Treating concentration trends as a first-order process is a hypothesis that needs substantiation. Examination of alternative measures of loadings trends needs to be undertaken.

6. Additional resources and efforts need to be expended on source identification projects for Great Lakes loadings. There are relatively few studies that have been done or which are underway or planned (e.g., Hites, Cohen, Holsen, Bidleman). There are no IADN source identification projects on the horizon, other than the PSCF analysis by R. Hites (IU) and his students recently submitted for publication at least as funded by the GLNP. It is not obvious how the relative significance of out-of-basin sources should be assessed. Atmospheric trajectories are highly variable, thus selection of averaging time is critical. The information ultimately required is annual source-to-receptor quantities, but this may vary from year to year and certainly season to season as a result of variability in climatology and source strength variability.

7. The nomination process for new chemicals is not working. IADN needs to undertake a new and more open process for actively seeking recommendations for listing and de-listing chemicals.

8. As stated in the last review report, IADN is still “a best kept secret” operating in the open. The US EPA ORD should be brought into the picture by convincing them of the national and international value of IADN for their efforts. The publications of IADN are unbalanced with most coming from one US laboratory. Whereas the the significant and successful efforts of R. Hites (Indianan University) should be warmly applauded, there has not been a corresponding effort by Canadian scientists to publish results and achieve the same level of visibility at scientific meetings. Making the data more web-accessible, as noted above, may improve the visibility and utility of the program.

9. The IADN Steering Committee, while taking justifiable pride in its accomplishments, needs to be more self-critical of its short comings. If there are insufficient data to make a credible loadings estimate, this should be presented as

such to the public. Uncertainties in the measurements and loadings estimates should be clearly and honestly presented.

10. The International Joint Commission (IJC) has over the years played a significant role in enhancing US-Canada cooperation and promoting joint scientific investigation of the shared Great Lakes. The Review Panel understood that the IADN Steering Committee increasingly views the IJC as irrelevant in this context. Given the strong tradition of public support for the IJC and its many accomplishments this change in attitude is very regrettable. More involvement of the IJC is desirable.

Comments on the big picture....

There are numerous loadings pathways for toxic chemical entry into the Great Lakes, and it is important, first, to understand their relative importance for each chemical of concern in each Great Lake. The development of these credible mass balance estimates for each compound of concern in each Great Lake is a critically important management and scientific tool, and is essential to developing sound policy to ameliorate the contaminant problems in the lakes. At the present time such estimates are not available for many chemicals of concern in one or more lakes.

As a first step, a project should be undertaken to assemble all data relevant to the mass-balance question for each compound of concern for each lake, perhaps similar in scope to the Strachan and Eisenreich Report of 1988 but improved in application of new knowledge and processes. The data should then be used estimate the relative importance of each pathway. In cases where data are not available and an independent "scientific judgment" cannot be credibly made, targeted measurements should be identified and made. This may involve, for example, the measurement of particular compounds in key tributaries to a given lake, or the measurement of a given compound in the atmosphere that has heretofore been unmeasured in the Great Lakes region.

From a policy perspective, the mass balance estimates often only need to be qualitatively correct – exact answers are likely not scientifically achievable or affordable, and will be unnecessary from a policy and management perspective.

For any significant loading pathway (for a given chemical to a given lake) it is also important to identify and determine the relative importance of different sources. Only then are concerted efforts possible to reduce loadings and improve water quality. The development of such source-receptor information cannot generally be achieved practically by monitoring and surveillance alone, but will require modeling tools to be combined the measurements.

It is recognized that the atmospheric deposition pathway is particularly complex in this regard. Depending on the pollutant, different strategies to combine monitoring and modeling analyses to develop source-receptor details should be made. For compounds for which reasonably accurate and complete emissions inventories can be made, a

comprehensive fate and transport model may be able to be used. For compound with no emissions inventory, back-trajectory based approaches (e.g., PSCF analysis) may be used to develop hypotheses regarding the relative importance of different source regions, and receptor-based approaches can be used to develop information on the relative importance of different source categories.

The relative importance of different loadings pathways and sources contributing to a given pathway may change over time, and so periodic updates should be made to ensure that information remains relevant.

In essence the Panel views the IADN program as an *essential* and *successful* component of the management of the Great Lakes water quality. Its full benefits cannot, however, be realized without strengthening the other components, notably the identification of all relevant sources and fate processes.

Other comments

There are uncertainties in the algorithm used to estimate loadings, and it would be useful if the relative magnitudes of the different sources of uncertainty for each compound for each lake could be estimated (e.g., Hoff publication in JGLR). Then, there are uncertainties associated with the use of the algorithm itself (e.g. difficulty in including the "urban" effect and effects other spatial variations). While it may not be possible to make quantitative estimates of these additional, more fundamental uncertainties, they should at least be more formally acknowledged, and attempts made to compile at least qualitative estimates of their magnitude should be made.

The 1 day in 12 sampling strategy may not be adequate for all compounds, including for example, current use herbicides and pesticides, due to potentially large temporal variations in the atmospheric concentrations of these compounds. The 1 day in 12 sampling strategy should be re-examined for existing compounds in the network to determine its adequacy for estimating average concentrations.

Non-detects: loadings analysis should simply include the range of possible answers based on the possible range of concentrations from zero up to the detection limit.

The Steering Committee sought answers to the following questions on the implementation of IADN:

1. **Has IADN been successful in meeting the goals for monitoring and surveillance set out in the Great Lakes Water Quality Agreement Annex 15 and the Clean Air Act Section 112 while striving to remain current?**

Answer: In total NO, but in parts, YES

a. Annex 15 declares that IADN is a research, monitoring and surveillance network, with the product being evaluation of atmospheric concentration trends and loadings to the Great Lakes Basin Ecosystem. IADN is presently considered by the Parties as only a Monitoring and Surveillance network.
b. To reduce uncertainties in loadings, understanding and quantification of deposition processes and parameters requires significant improvements.
c. Toxic chemical loading, as stated in the Annex 15 and CAA Sec 112, should be considered for the Great Lakes Basin Ecosystem, not just directly to the Great Lakes.
d. The effort for IADN to remain current includes improving methodologies for sampling and analysis, AND improvement in concentration trends, loadings and source identification. IADN should consider trace metals and Hg as candidate analytes.
e. IADN is successful in monitoring air concentrations but not successful in reducing the uncertainty in loadings estimates.

2. **Did IADN do what it said it would do in IP2? Were the right questions asked in IP2 or did more important scientific questions go unanswered? Were IADN's priorities correctly placed? Were the right measurements being made? Were the right chemicals being measured?**

IP Goal 1: Answer: NO

a. Target chemical loadings directly to the Great Lakes are considered in IADN but loadings to the Basin are not. There has been inadequate improvement in loading estimates and an adequate degree of confidence needed for loadings in IP3 has not been specified.
b. Uncertainties in loadings to lakes have not been adequately captured– e.g., urban influence
c. Given the present availability of resources, IADN should focus on estimating only downward fluxes. Upward volatilization should be dealt with separately and viewed as only one of several loss processes including degradation, outflow and sediment burial. It is fundamentally undesirable to combine a fairly accurate estimate of downward fluxes with a very inaccurate estimate of upward fluxes yielding an inaccurate and uncertain estimate of net transport. Estimates of volatilization require accurate water concentrations which regrettably are not available for the same location and times as the deposition data. Further a research effort is required to evaluate net air-water exchange fluxes using innovative methods such as fugacity gradients.

d. Volatilization should be dealt with separately based on availability of water data. (try new techniques at a research water site to evaluate net air-water exchange flux, fugacity gradients).

IP Goal 2: Answer: Yes

a. Quality air concentrations are being determined, and consistency and continuity has been achieved. The reason for systematic differences between PCB air concentrations of Canadian and US laboratories must be determined and corrected. Canadian rain concentrations of PCBs high by 10 x or so should be resolved or the procedures abandoned.

IP Goal 3: Answer: NO Help determine sources of continuing inputs of chemicals

a. The Parties should apply regional air transport and deposition models with air concentrations to identify and quantify continuing sources.

b. Urban-remote or urban-background studies have proved valuable. A strategy should be developed to analyze and use these data? Urban sites may be politically necessary to profile the urban contamination in the atmosphere to the public. How best to raise the public profile and support for IADN may be enhanced by focusing on these urban activities? (IADN Day in Major Great Lake cities?)

3. Was the work scientifically credible? What was the overall quality of the work being conducted under the IADN program?

Monitoring. How does IADN compare against the quality and standards of international networks and campaigns to monitor and assess the deposition of POPs?

Answer: YES Partly

IADN is a flagship international network of long duration and utility. It is generally comparable to EMAP although it did not include trace metals and POPs before 2000. EMAP does include most all the POPs on IADN's list. Analyses are done by a few laboratories with a strong QA effort; NJADN (New Jersey Atmospheric Deposition Network) includes all the POPs on IADN list and also includes trace metals, OC/EC, N, P and Hg (ppt). NJADN also makes strong use of the air monitoring and strategic research in linked field campaigns to address key questions on process, sources and fate. EMAP includes emissions, measurements, and modeling.

Research (Extensions to IADN). Were the research projects scientifically sound? Innovative? Timely? Do they enhance or build upon IADN? Do they address research areas listed in Annex 15 of the GLWQA?

Research activities linked to IADN have resulted in many important publications and provide significant added value to the IADN. Further encouragement to researchers to build upon the IADN platform are encouraged including funding from the GLNP.

Improvements of methods have resulted in significant enhancement to the IADN product. One of the Master stations might be utilized to obtain frequent water concentrations for loadings estimates.

Were the rate and mechanism of delivery of the information to the scientific community, to policy makers, and to the public reasonable? Do IADN's current methods of distributing information (web site, Loading reports, press releases, journal articles) meet the needs of the scientific community, policy makers, and the public? What other mechanisms should IADN be using?

Data (concentrations) delivery takes far too long from sample collection to reporting to be useful in scientific and policy-making areas. The target should be 6-8 months.

Data should be released on a regular basis and independent of the release in loading estimates.
Web-based data delivery should be emphasized including linkages to/on other relevant sites in US, CAN, and Europe.

Scientific output (publications; conference presentations) of IADN is now excellent although unbalanced between Parties. Canada should increase its scientific profile relative to IADN, perhaps by MSC soliciting academic involvement.

IADN should make use of the press as a medium to disseminate IADN information; IADN needs a more effective way to get the message out. This will require a concerted effort by all parties.